

URANIUM DIFFUSION

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RESEARCH OBJECTIVES

Transport of uranium (U) in contaminated soils and sediments is of great concern. Environments affected by U contamination can be very challenging to understand because extreme disequilibrium is likely in such environments, especially during early stages of waste migration, when solutions are still often either highly acidic or highly alkaline. The impact of diffusion is especially important when large fractions of the subsurface have low hydraulic conductivities relative to a small fraction of interconnected preferential flow paths, and where hydraulic gradients are low. In this study, we are concerned with diffusion of U(VI) species, since these are commonly much more soluble and mobile than U(IV) species. Because U(VI) forms a variety of strongly pH-dependent solution complexes and surface complexes, its sorption—and thus its mobility—is strongly pH-dependent. Experiments are being conducted to test various models for predicting transient U(VI) diffusion.

APPROACH

Measurements were obtained on U(VI) diffusion into two different soil types, one neutral and the other slightly alkaline. To each of these soil types, we applied U(VI) solutions, one acidic and the other alkaline. Periodic measurements of redox potential profiles within the soil columns, and of pH and U concentrations in the boundary reservoirs, were obtained. Profiles of the total U and U(VI) distribution in the sediment column were obtained by micro- x-ray absorption near-edge structure (micro-XANES) spectroscopy. Measured U profiles were compared with various model predictions.

ACCOMPLISHMENTS

Micro-XANES analyses confirm that U remains in oxidized U(VI) forms within systems having low organic carbon availability. Strong pH-dependent apparent U(VI) diffusivities are obtained in transient experiments. These apparent diffusivities are in fair agreement with predictions based on aqueous-phase U(VI) diffusivities, soil porosities, and pH-dependent sorption isotherms. Ongoing sorption studies (Zheng et al., 2003) are being conducted through a related NABIR-supported project.

SIGNIFICANCE OF FINDINGS

Predicting the mobility of U in soils and sediments requires an understanding of its diffusive transport. Such understanding

is being provided in this study, through direct measurements of U(VI) diffusion profiles and comparisons with U(VI) sorption isotherms.

RELATED PUBLICATIONS

Tokunaga, T.K., J. Wan, T.C. Hazen, E. Schwartz, M.K. Firestone, S.R. Sutton, M. Newville, K.R. Olson, A. Lanzirotti, and W. Rao, Distribution of chromium contamination and microbial activity in soil aggregates. *J. Environ. Qual.* 32, 541–549, 2003.

Zheng, Z., T.K. Tokunaga, and J. Wan, Influence of calcium carbonate on sorption of U(VI) to soils. *Environ. Sci. Technol.*, 2003.

ACKNOWLEDGMENTS

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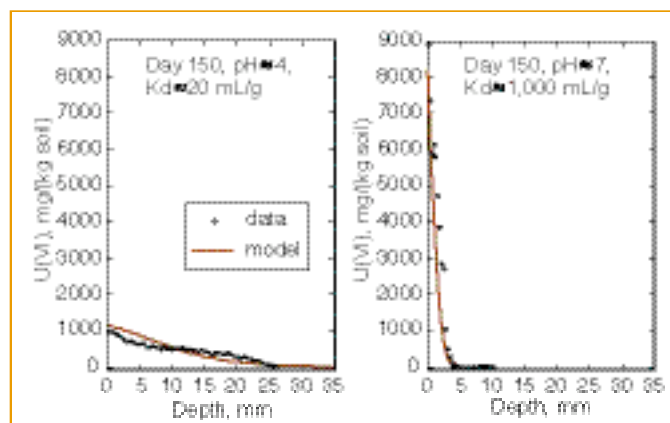


Figure 1. Micro-XANES profiles at day 150, for U(VI) diffusing into Oak Ridge soil from initially acidic (left) and initially alkaline (right) solutions. Neutralization of the alkaline system resulted in strong U(VI) sorption and retardation of the diffusion front. Model calculations are based on measured porosities, aqueous-phase U(VI) diffusivities, and pH-dependent U(VI) sorption.